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(54) CURABLE COMPOSITION

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a curable composition not causing stringiness in use, having a small slump in an uncured state, excellent weather resistance for a long period of time after curing and a sufficiently high degree of lightening.

SOLUTION: This curable composition is characterized by comprising an oxyalkylene polymer containing a reactive silicon group represented by general formula (1) -SiXaR1(3-a)...(1) [R1 is a monofunctional organic group; X is a hydroxy group or a hydrolyzable group; a is 1-3] and a polymer containing a (meth)acrylic acid alkyl ester monomer unit and minute hollow bodies of organic resin.

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CLAIMS

Claim 1]an oxyalkylene polymer (A) which has a reactive silicon group expressed with a following general formula (1), and -SiX₃R¹ (3-u) . (1) [Claim(s)]

show a hydroxyl group or a hydrolytic basis among [typa, and a shows an integer of 1-3, respectively. However, when R1 may be the same respectively or it may differ, when two or mora R1 exist, and two A univalent organic group of the carbon numbers 1-20 in which R1 may have a substituent, and X

Meta) A polymer (B) including an acrylic-acid-alkyl-ester monomeric unit. or more X exists, X may be the same respectively or may differ.] A hardenability constituent comprising.

An organic resin minute hollow body (O).

[Olaim 2]The hardenability constituent according to claim 1, wherein said oxyalkylene polymers (A) are 6000 or more number average molecular weight distribution Claim 3]The hardenability constituent according to claim 1 or 2 being a polymer cheracterized by (Mw/Mn) oxyalkylene polymer.

An acrylic-acid-alkyl-ester monomeric unit whose carbon number of an elkyl group is ten or more A carbon number of an alkyl group of said polymer (B) is an acrylic-acid-alkyl-ester (meta) monomeno unit of 1-8 as an acrylic-acid-alkyl-ester (meta) monomeric unit. comprising the following:

[Olaim 4]The hardenability constituent according to any ona of claima 1 to 3 said polymer's (B's)'s being a polymer including an acrylic-acid-alkyl-aster (mota) monomeric unit, and baing a polymer Claim 5]The hardenability constituent according to any one of claims 1 to 4, wherein said organic reain minute hollow body (C) is a polyvinylidane chloride system resin minute hollow body. which has a reactive silicon group expressed with the above-mentioned general formula (1).

[Trenslation done.]

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DETAILED DESCRIPTION

Detailed Description of the Invention

hardenability constituent suitable as the main ingradients, such as sealant and adhesives, in more [Field of the Invention] This invention relates to a hygroscopic-surface-moisture hardening type detail about a hardenebility constituent.

likoxy silyl groups, (it is hereafter called a 'hardened type oxyalkylene polymer'.). It can harden slso Description of the Prior Art The oxyslkylene polymer which has reactive silloon groups, such as it a room temperature with the hygroscopic surface moisturs in the air, and since the hardened

property over various adherends, it is widely used as sealant, adhesives, base resin of the constituent naterial which has rubber elasticity is obtained and after hardening is excellent also in the adhesiva or covering / seal, etc.

0004]As a method of raising the weatherability of a hardened type oxyalkylene polymer, For example, applied, for example to sealant, under the exposure condition in the outdoors, a white blush mark and s crack may arise on the hardened material surface, and improvement in weatherability is called for. 0003]However, when such a hygroscopic-surface-moisture hardcning type oxyalkylene polymer is the method indicated by JP,59-78223,A, JP,59-122541,A, JP,60-31556,A, JP,63-112642,A, JP,6-172631.A, etc. is known. According to these gazettes, it is supposed by making a hardened type

exyalkylene polymer contain the acrylic-acid-alkyl-ester system polymer which has a reactive silicon 0005]To the hardened type oxyalkylene polymer containing the acrylic-acid-alkyl-ester system group (meta) that improvement in weatherability can be aimed at.

colymer which has a reactive silicon group (meta). The presentation which furthermore added bulking sgents, such as glass balloons, is indicated by UP.5-86325.A as an axample, and is indicated [that a weight saving, saving resources, rust prevention vibration proof, etc. are reslizable and] by by using the presentation concerned as a costing agent for vehicles.

point of a weight saving, there was a problem that it was [that it is easy to produce a cobwebbing at the time of use] inferior to workability. When it was especially used for a joint part as sealant, there Problem(s) to be Solved by the Invention However, when it used for the use of sealant, adhesives, eto. in addition to the presentation of an indication being still insufficient for JP,5-86325A in the was a problem that a slump (lappet) was large.

1000] This invention is made in view of the problem of the above-mentioned conventional technology. siso in the weatherability over the long period of time after hardening, and it is that the degree of a weight saving also provides a hardenability constituent high enough further. * does not occur, but the slump of the purpose at the time of un-hardening is small, is excellent

polymer as a result of repeating research wholeheartedly that this invention parsons should attain the sbove-mentioned purpose, it found out that cobwebbing prevention which was not sble to be attained, reduction of a slump, weathersbility, and all the four characteristics of a weight saving could added to a hardened type oxyalkylene polymer containing an acrylic-acid-skyl-ester (meta) system Means for Solving the Problem]By using an organic resin minute hollow body as a bulking agent

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oe attained in a presentation of an indication in the above-mentioned gazette, and this invention was

However, when R1 may be the same respectively or it may differ, when two or more R1 exist, and two 0009]Namely, an oxyalkylone polymer (A) in which a hardenability constituent of this invention has a show a hydroxyl group or a hydrolytic basis among [type, and a shows an integer of 1-3, respectively aorylic-acid-alkyl-oster monomeric unit and an organic resin minute hollow body (C) are included. [0010]As for an oxysikylene polymer (A) in this invention, it is preferred that they are 6000 or more A univalent organic group of the carbon numbers 1-20 in which R¹ may have a substituent, and X or more X exists, X may be the same respectively or may differ. A polymer (B) including] (meta) number average molecular weights and a 1.7 or less molecular weight distribution (Mw/Mn) reactive silicon group expressed with a following general formula (1) and -SiX_aR¹ (3-a) - (1) oxyalkylene polymer.

silicon group expressed with the above-mentioned general formula (1). As for an organic resin minute hollow body (C) in this invention, it is preferred that it is a polyvinylidene chloride system resin (meta) monomeric unit of 1-8 and an acrylic-acid-alkyl-ester monomeric unit whose carbon number preferred that a carbon number of an alkyl group is a polymer including an acrylic-acid-alkyl-ester including an acrylic-acid-alkyl-ester (meta) monomeric unit, and is a polymer which has a reactive 2011]A polymer (B) in this invention as an acrylic-acid-alkyl-ester (meta) monomeric unit. It is of an alkyl group is ten or more (meta). As for a polymer (B), it is preferred that it is a polymer

group provided with the hydroxyl group or the hydrolytic basis. As the hydrolytic basis concerned, for le, a halogen atom, an alkoxy group, an acyloxy group, an alkenyloxy group, an amino group, a Embodiment of the Invention] First, the oxyalkylene polymer (A) in this invention is explained. The oxyalkylene polymer (A) in this invention is an oxyalkylene polymer which has the reactive silicon KETOKISHI mate group, an aminooxy group, a carbamoyi group, and a sulfhydryl group can be minute hollow body.

especially a preferred methoxy group. R^1 is a univalent organic group of the carbon numbers 1-20, preferred not to have a substituent. As for the carbon number of R¹, it is preferred that it is 1-16, preferred, it is more preferred that it is 1-4, as such a basis, a methoxy group, an ethoxy basis, a propoxy group, and a butoxy group are mentioned, end the carbon number of an alkoxy group has even if the organic group concerned has a substituent, it is not necessary to have it but, and it is 0013]As the above-mentioned alkoxy group, the alkoxy group of the carbon numbers 1-6 is mentioned, and an alkoxy group is especially preferred. and it is more preferred that it is 1-8.

concerned may have straight chain shape and which a branched stats and annular structure. As such sn organic group, an elkyl group, a cycloalkyl group, an aryl group, and an aralkyl group are mentioned, for example. As an alkyl group, the alkyl group of the carbon numbers 1-4 is preferred, and a methyl group, an ethyl group, a propyl group, and a butyl group are mentioned as such an alkyl group. When 0014]When R1 is a univalent organic group which does not have a substituent, the organic group R1 is a univalent organic group which has a substituent, the kind in particular of the substituent concerned is not restricted.

[0015]in the reactive silicon group, when two or more basis or organic group R¹ expressed with X preferred that it is 2 or 3, and it is more preferred that it is 2. The basis which is not bulky as \mathbb{R}^1 is preferred, and a methoxy group is preferred as a hydrolytic basis. Therefore, especially in this construction of an oxyalkylenc polymer, as for the number (a in a general formula (1)) of X, it is invention, a methyl dincthoxy silyl group and a trimethoxysilyl group are preferred as a reactive exist, they may be the same respectively or may differ. From a viewpoint of promoting bridge

number of the reactive silicon groups per oxyalkylene polymer monad nor the binding site in particular of the reactive silicon group in an oxyalkylene polymer is restricted. That is, the oxyalkylene polymer [0016]The oxyalkylene polymer (A) in this invention has a reactive silicon group expressed with the general formula (1) explained above into the molecule of an oxyalkylene polymer. Here, neither the A) should just have the above-mentioned reactive silicon group one or more per monad, and the nttp://www4.ipdl.inpit.go.jp/cgi-bin/tran_web_ogi_ejje?atw_u=http%3A%2F%2F%2Fwww4.ipdl.i.. 2010/03/03

existence part may be both an end of an oxyallylene polymer, a side chain, an end and a side chain,

0017]The oxyalkylene polymar which has a reactive silicon group should just be a polymer which has oxyalkylena aa a repeating unit, and oxyalkylene may consist only of one sort and may consist of two compound as for an oxyalkylene polymer so that it may mention later, it may have a functional group of initiator origin in the molecule. Since the hydroxyl group to contain may be made to react to other functional groups and polymers quantification may be carried out, the oxyalkylene polymer may have or more sorts. Especially when a repeating unit consists of two or more sorts of oxyalkylene, Even if each oxyalkylene connects at random end forms the random polymer of oxyalkylene, oxyalkylene of mit in the molecule. Since it is common to make cyclic ether react to an initiator (initiator), and to en identical kind may collect and connect with blook like shape, and may form the blook copolymer. [0018] The oxyalkylene polymer may have chemical structure other than oxyalkylene as a repeating n the molecule combination (for example, urethane bond etc.) which a hydroxyl group and other unctional groups reacted and produced, so that it may mention later.

(0019]As for the oxyalkylene polymer (A) in this invention, it is preferred that it is what is obtained by using as a raw material the oxyalkylene polymer which has a functional group, and making a part or all general formula (1) react. As an oxyalkylene polymer which has a functional group for introducing a sactive silicon group, an oxyalkylene polymer with a hydroxyl group, an unsaturation group, and an of the functional group, and the compound which has a reactive silicon group expressed with a

etrahydrofuran, can be carried out, and it can be obtained. In this case, alkaline metal catalysts, such 0020]In this invention, it is preferred to use the oxyalkylene polymer (henceforth a "hydroxyl group and oxyalkylene polymer") which has a hydroxyl group at the end as an oxyalkylene polymer which univalent], or polyvalent carboxylic acid, as an initiator, Ring opening polymerization of the cyclic ass a functional group for introducing a reactive silicon group. A hydroxyl group and oxyalkylene as a potassium system compound and a caesium system compound, a composite metal cyanide polymer uses active hydrogen containing compounds, such as univalent, [polyhydric alcohol or ether, such as ethylene oxide, propylene oxide, 1,2-butylene oxide, 2,3-butylene oxide, and a complex catalyst, and a metalloporphyrin catalyst can be used as a polymerization catalyst. sooyanate group as a functional group is mentioned.

ydrogen, in this invention, since the hardenability constituent which is excellent in the pliability after active hydrogen containing compounds which have 2-6 active hydrogen. As for the number of active quentification can be carried out (JP,82-240320,A). On the other hand, as a polymerization catalyst, nardening and an adhesive property is obtained, it is still more preferred that it is 2 or three pieces. exyeligiene polymer. Since the polymer of low molecular weight is obtained comparatively, terminal 0021]It is preferred to use the multivalent active hydrogen containing compounds which have 2-8 '0022]When an alkaline metal catalyst is used as a polymerization cetalyst of a hydroxyl group end when a composite metal oyanide complex catalyst is used, the polymer of the amount of polymers active hydrogen as the above-mentioned initiator, and it is more preferred to use the multivalent salogenated compounds, such as a methylane chlorida, react, it can quantify many and polymers nydroxyl groups of the polymer concerned is made into sodium alcoholate atc., by making many As multivalent active hydrogen containing compounds, polyhydric alcohol is preferred. and narrow molecular weight distribution can obtain (JP,3-72527,A).

solymer, in this invention, it is preferred that it is a composite metal cyanide complex catalyst. The complex which uses zinchexacyano cobaltate as the main ingredients as a composite metal cyanide (0023)As for the polymerization catalyst used in order to obtain a hydroxyl group end oxyalkylene

[0025]By using the hydroxyl group end oxyalkylene polymer explained above, the oxyalkylene polymer group end oxyalkylene polymers may be two or more sorts of mixtures in which a molecular weight differs from chemical structure, and adjustment of the physical properties after hardening and the [0024]As a hydroxyl group end oxyalkylene polymer, the polyoxypropylene polyol of 2 - 6 value is preferred, and polyoxypropylene diol and polyoxypropylene triol are especially preferred. Hydroxyl (A) which has a reactive silicon group by following (I) - the method of (IV) can be obtained, for adjustment of the hardening characteristic of them are attained by using such a mixture.

(1) A method to which the unseturation group concerned and hydrosily! compound are made to react

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IP,2002-155201,A [DETAILED DESCRIPTION]

after changing the terminal hydroxyl groups of a hydroxyl group end oxyalkylene polymer into an

(II) A method to which a hydroxyl group and oxyalkylene polymer and an isocyanate group content compound are made to react after changing the terminal hydroxyl groups of a hydroxyl group end (III) A method to which the isocyanate group concerned and an active hydrogen content silicon oxyalkylene polymer into an isocyanete group. silicon compound are made to react.

concerned after changing the terminal hydroxyl groups of a hydroxyl group end oxyalkylene polymer (VI) How to make a suifhydryl group content silicon compound react to the unsaturation group nto an unsaturation group.

exyalkylene polymer obtained by the above-mentioned method, and introduce an unsaturation group 0026](0 In a method, make the compound which has a hydroxyl group, a reactant functional group. into the end of an oxyalkylene polymer first, for example, Subsequently, the reactive silicon group compound expressed with a following general formula (2) react to this polymer (hydrosilylation expressed with a general formula (1) can be introduced by, for example, making the hydrosilyl and an unsaturation group react to the terminal hydroxyl groups of the hydroxyl group end

1SiX, R (3-1) - (2)

0027JAs the above-mentioned unsaturation group, an alkenyl group, an acrylyl group, a methacryloy group, etc. are mentioned. As a hydroxyl group and a reactant functional group, a halogen atom, a R1, X, and a are synonymoue with eaid R1, X, and a among [type.]

ond are formed, respectively. For example, when the hydrosilyl compound expressed with the aboveoxyalitylene polymer into an allyloxy group, terminal hydroxyl groups turns into a basis expressed with unctional groups react to terminal hydroxyl groups, an ether bond, an ester bond, and a urethane mentioned general formula (2) is made to react after changing the terminal hydroxyl groups of an carboxyl group (or halo formyl group), an isocyanate group, etc. are mentioned, and when these

a following general formula (3). -0-C3H6-SiXaR1 (3-a) - (3)

reactive silicon group expressed with the end of an oxyalkylene polymer by a general formula (1) via a socyanate group content silicon compound react, As shown in a following general formula (4), the .0028](II) In a method, for example by making a hydroxyl group end oxyalkylene polymer and an R1, X, and a are synonymous with said R1, X, and a among [type.]

urethane bond can be introduced. - OCONH-R2-SIX, R1 (3-2) - (4)

R² shows the divalent hydrocarbon group of the carbon numbers 1−20 among [type. R¹, X, and a arc [0029](III) In a method, for example, make a hydroxyl group end oxyalkylene polymer and a synonymous with said R1, X, and a.]

becomes larger than the number of hydroxyl equivalents, and change the terminal hydroxyl groups of following general formula (6) can be introduced by, for example, making the active hydrogen content a hydroxyl group end oxyalkylene polymer into an isocyanate group first. Next, the reactive silicon group expressed with the end of an oxyalkylenc polymer by a general formula (1) as shown in a polyisocyanate compound react on the conditions to which an isocyanate number of equivalent silicon compound expressed with a following general formula (5) react to this polymer.

W-R2-SIX₈R1 (3-s) - (5)

W shows among [type the active hydrogen containing group chosen from the group which consists of a hydroxyl group, a carboxyl group, a sulfhydryl group, the 1st class amino group, and the 2nd class amino group. R², R¹, X, and a are synonymous with said R², R¹, X, and a.] - NH-W1-R2-SIX,R1 (3-2) - (6)

W1 shows among [type the divalent basis chosen from the group which consists of -COO-, -CO-, -COS-, -CONH-, and -CONW2-, However, W2 shows a univalent organic group and R2, R1, X, and a are synonymous with said R2, R1, X, and a.] nttp://www4.ipdl.inpit.go.jp/ogf-bin/tran_web_ogi_ejje?atw_u=http%3A%2F%2Fwww4.ipdl.i. 2010/03/03

outent silicon compound expressed with this polymer and a following general formula (7) react of a statement in a method at the above-mantioned (I) using a hydroxyl group end oxyalkylone polymer, for example. Subasquantly, the reactive silicon group expressed with the end of an

R², R¹, X, and a are synonymous with said R², R¹, X, and a among [type.] HS-R2-SiX,R1 (3-2)-- (7)

shove-mentioned general formula (7) is made to react after changing the terminal hydroxyl groups of an oxyalitylene polymer into an allyloxy group, terminal hydroxyl groups turns into a basis expressed 0031](VI) In a method, when the sulfrydryl group content silicon compound expressed with the vith a following general formula (11).

R1, X, and a are synonymous with said R2, R1, X, and a among [type.] -0-C3H6-S-R2-SIX3R1 (3-3)...(8)

0032]The method of the above-mentioned (I) is the method of transforming the unsaturation group soncemed, after using terminal hydroxyl groups of a hydroxyl group and oxyalkylene polymer as an unsaturation group, but the following methods are possible for it as a modification mode of this j083]Namely, in the case where carry out ring opening polymerization of the cyclic ether, and a ydroxyl group end oxyslkylana polymer is obtained under existence of an initiator. As cyclic ether, by using together unsaturation group content monoepoxide, auch as allyl glycidyl athar, glycidyl acrylato, and glyoidyl methacrylate, An unsaturated bond can be introduced into the side chain of a hydroxyl

oxyalkylene polymer which has a reactive silicon group expressed with an end and a side chain by the shain by changing the terminal hydroxyl groups of this hydroxyl group and oxyalkylene polymer into 0034]Since an oxyalkylene polymer with an unsaturation group is obtained by an end and the side an unsaturation group like the method of the above-mentioned (I), The unsaturation group in the olymer concerned can be changed by the same method as the above-mentioned (I), and the above-mentioned general formula (1) can be obtained. group end oxyalkylene polymer (JP,3-79627.A).

group and terminal hydroxyl groups, such as an allyf end polyoxypropylene monocar, can obtain. After unsaturation group, for example as active hydrogen containing compounds which are the initiators in the case of obtaining an oxyalkylene polymer. The oxyalkylene polymer which has an unsaturation polymer as an unsaturation group, by the still more nearly same method as the above-mentioned (I). nentioned (0. Namely, by using a compound with an active hydrogen group like allyl alcohol, and an changing the terminal hydroxyl groups of the polymer concerned into an unsaturation group by the 0035]The method of further the following is also possible as a modification mode of the abovesame method as the above-mentioned (I) and using all the functional groups of an oxyalkylone The reactive silicon group axpressad with the above-mentioned general formula (1) by the

CH,=CR3COOR4 (9)

10036]The oxyalitylene polymer (A) explained above forms a hardonability constituent with a polymar (B) and organic resin minute hollow body (C) including an acrylic-acid-alkyl-aster (meta) monomeric that it may mention later. In such a use, in order to have to apply to adherend before a hardenability unit. This hardenability constituent can be used as a raw material of sealant or elastic adhesives so constituent hardens, moderate workability is required and it is preferred that the viscosity in a room temperature is low for that purpose. As for after hardening, it is preferred to have rupture stress moderate as sealant or elastic adhesives and the degree of breaking extension.

exvalkylene polymer can be introduced.

this invention, it is preferred in 25 ** that it is below 30 Pa and s. Although the minimum in particular [0037] Therefore, as for the viscosity of the viewpoint of workability to an oxyalkylene polymer (A), in of viscosity is not restricted, 1 or more Pa-s is preferred, and 5 or more Pa-s is more preferred. 0038]As for an oxyalkylene polymer (A), in this invention, it is preferred that they are more than number average molecular weight (Mn) 6000 and a 1.7 or loss (namely, 1.0-1.7) molecular weight When the viscosity of an oxyalkylene polymer (A) exceeds 30 Pa-s, it is in the tendency for the vorkability of the hardenability constituent obtained to worsen.

nttp://www4.ipdl.inpit.go.jp/cgi-bin/tran_wab_ogi.ejje?atw_u=http%3A%2F%2Fwww4.ipdl.i. 2010/03/03 om the rupture stress of a hardened material, and a viewpoint of the degree of breaking extension

liatribution (Mw/Mn) oxyalkylene polymer. As for Mn of an oxyalkylena polymar (A), it is preferred

IP,2002-155201,A [DETAILED DESCRIPTION]

0039]As for Mw/Mn of an oxyalkylene polymer (A), it is more preferred that it is 1.6 or less, and it is scalant or elastic adhesives, it is in the tendency for workability, such as extrusion nature, to fall. On still more preferred that it is 1.5 or leas. When Mw/Mn of an oxyalkylane polymer (A) exceeds 1.7, it is in the tendency for the cure rate of the hardenability constituent obtained to fall. This is based on 1.7 or less thing with the thing in which Mw/Mn exceeds 1.7, since that in which Mw/Mn exceeds 1.7 has large molecular weight distribution, there is much content of a low molecular weight constituent, Namely, an average molecular weight is the same oxyalkylene polymer (A), When Mw/Mn compares molecular weight distribution whose value of Mw/Mn is 1.7 or less can be obtained by, for example that it is 6000–50000, and it is more preferred that it is 8000–30000. When using the hardenability the other hand, the hardenability of a constituent tends to be infarior when Mn ia less than 6000. as for an oxyalkylene polymer (A), a cure rate becoming slow when the molecular weight is small. therefore it is in the tendency for a cure rate to fall. The narrow oxyalkylene polymer (A) of the constituent which viscosity becomes high, for example, is obtained when Mn exceeds 50000 as using the composite metal eyanide complex mentioned above as a cyclic ether ring opening

before hardening about Mn of an oxyalkytena polymer (A). In this invention, Mw/Mn is the value which 0040]Mn in this invention means the number average molecular weight of the styrene conversion measured by gel permeation chromatography (GPC) with a tetrahydrofuran solvent, and means Mn re(ed) Mw (weight average molecular weight) of the styrene conversion measured by GPC with a tetrahydrofuran solvent from Mn (numbar average molecular weight) massured on the same polymerization catalyst.

usually polymerizing the unsaturation group content monomer which uses an acrytic-acid-alkyl-ester monomer. it is a compound which has an unsaturated bond (preferably carbon-carbon double bond). alkyt-ester monomeric unit in this invention (meta). (Meta) The polymer which has a repeating unit which consists of acrylic acid alkyl ester is meant, and the polymer concerned can be obtained by the compound which can form a polymer is said, and acrylic acid alkyl ester (meta-) means acrylic nardenability constituent of this invention is explained. With a polymer including the acrylic-acidmeta) monomer as an essential ingredient. In this invention, with an unsaturation group content 0041]Next, a polymer (B) including the acrylic-acid-alkyl-ester (meta) monomeric unit in the 0042]The acrylic-acid-alkyl-ester monomer in this Invention (meta) can be expressed with a acid alkyl ester or methacrylic acid alkyl ester. following general formula (9).

0043]Although R⁴ in a general formula (9) is an alkyl group, in this invantion, a substituted alkyl group with which at least one of tha hydrogen atoms of an alkyl group was replaced by hydrocarbon groups, monomer concerned, or two sorts or more, and the kind or number of repeating units in a polymer (B) preferred that the carbon number of an alkyl group is a polymer including the acrylic-acid-alkyl-estor such as cyclic hydrocarbon groups, shall also be included in an alkyl group like an aralkyl group and a (meta) monomeric unit of 1–8 and the acrylic—acid—alkyl—ester monomeric unit whose carbon number of an alkyl group is ten or more (meta). That is, as for a polymar (B), it is preferred to have both the are not restricted in a limitation including the repeating unit which consists of an acrylic-acid-alkylrepeating unit which the carbon number of an alkyl group becomes from the acrylic-acid-alkyl-ester repeating unit which consists of one sort of unsaturation group content monomers other than the sorylic-acid-alkyl-ester monomers (meta) which are expressed with the above-mentioned general formula (9), or two sorts or more. One sort of acrylic-acid-alkyl-ester monomers or two sorts or [0044]Even if the polymer (B) in this invention has a repeating unit which consists of one sort of ester (meta) monomer. As for the rate of the acrylic-acid-alkyl-ester (meta) monomer in a total (0045]A polymer (B) in this invention as an acrylic-acid-alkyl-ester (meta) monomeric unit, it is more (meta) which are expressed with the above-mentioned general formula (9), It may have a monomer, it is preferred to exceed 50 mass %, and it is preferred. [of more than 70 mass %] (meta) monomer of 1-8, and the repeating unit which consists of an acrylic-acid-alkyl-eater 4s for R3, a hydrogen atom or a methyl group, and R4 show an alkyl group among [type.] cycloalkyl alkyl group. The carbon number in particular of an alkyl group is not restricted.

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nonomer whose carbon number of an alkyl group ia ten or more (meta). (Meta) It is in the tendancy

alkyl-ester monomer whose carbon number of an alkyl group is ten or more (meta), The acrylic-acidalkyl-aster (meta) monomer of 10-30 has a more preferred carbon number of an alkyl group, and the iorylic-acid-alkyl-ester (meta) monomer of 10-22 has a still more preferrad carbon number of an mprove by using such a combination as an acrylic-acid-alkyl-eater monomer. As an acrylic-acid-

or example, methyl acrylate (meta), ethyl acrylate (meta), acrylic acid (meta) propyl, butyl acrylate (meta), scrylic acid (meta) hexyl, scrylic scid (meta) octyl, 2-ethylhexyl scrylate (meta), and scrylic 0046]The carbon number of an alkyl group as an acrylic-acid-alkyl-ester (meta) monomer of 1–8,

meta), For example, decyl acrylate (meta), acrylic acid (meta) dodecyl, (Meta) Acrylic acid tetradecyl, 0047]As an acrylic—acid—alkyl—ester monomer whose carbon number of an alkyl group is ten or more icrylic scid (meta) hexadecyl, acrylic scid (meta) octadecyl, eicosanyl sorylate (meta), scrylic scid icid (meta) benzyl are mentioned.

sster (meta) monomer of 1-8, and the acrylic-acid-alkyl-ester monomer whose carbon number of an ormor/latter, it is preferred that it is 95 / 5 - 40/60 in a mass ratio, and it is more preferred that it (0048) in the case where the carbon number of an alkyl group uses together the acrylio-acid-alkylalkyl group is ten or more (meta), Although the ratio in particular is not restricted, as for the meta) docosa nil, and acrylic aoid (meta) hexacosa nil are mentioned.

ormulas (10) is mentioned as unsaturation group content monomers other than an acrylic-acid-alkylother than the monomeric unit concarnad other than an scrylio-acid-akyi-ester monomeric unit, as 0049]Tha polymer (B) in this invention may include unsaturation group content monomeric units nentioned above (meta). (Meta) Although the compound expressed with the following general aster monomer, it is not limited to these.

dispersibility of the polymer (B) in an oxyalkylene polymer (A) can also be raised. It is also considered in the middle of a polymerization at an oxyalkylene polymer (A), in such a case, a graft polymerization 0056]In this invention, it is preferred that a polymer (B) is a polymer including the acrylic-acid-alkyl-

B) under existence of an oxyalkylena polymer (A), mixed time and effort can be saved and the

that a part of unsaturation group content monomer for polymers (B) carries out graft polymerization sater (meta) monomerio unit mentioned above, and it is a polymer which has a reactive silicon group

thing functions as a compatibilizer and the dispersibility of a polymer (B) improves more.

CH,=CR3COOR5 _ (10)

त⁵ shows the univalent organic group or hydrogen atom except an alkyl group among [type. R³ is

etc. which contain the alkyl group, the glycidyl group, and polyalkylene glycol which have substituents univalent organic group except the alkyl group defined in a general formula (9). The univalent basis for example, a halogen atom, a hydroxyl group, an isocyanate group, a phenoxy group, a furfuryl 0050]The univalent organic group except the alkyl group in a general formula (10) means the synonymous with said R³.]

isocyanate alkyis, such as acrylate. (mata-) acrylate; --- phonoxy alkyi (meta-) acrylate L such as 2-phenoxyethyl (meta-) acrylate,]: — furfuryl (hydrogenation) groups, auch as furfuryl (meta-) sorylate ester (meta-) of polyalkylene oxide monecars, such as methoxy polyethylene-glycol (meta-) acrylate. Frimethoxysilane etc. (meta-) acryloxyalkyl alkoxysilanc; --- glycidyl (meta-) acrylate; --- the acrylic group, a reactive silicon group, etc.) other than a hydrocarbon group as such a univalent organic group are mentioned. As a compound exprassed with a general formula (10), (Meta) Aorylic acid, and tatrahydrofurfuryl (meta-) acrylate. . Have. (Meta) Acrylate; gamma-, (Methacryloxypropyl) Hydroxyethyl, (Meta) Hydroxyalkyl, such as acrylate. (Meta) Acrylate; Isocyanstoethyl. (Meta)

(meta) acrylamida, such as NN-dinethylacrylamide; Vinyl glycidy) ether, Unsaturation glycidyl ether, such as allyl glycidyl ether and methallyl glycidyl ether. Crotonic acid glycidyl, Monoslkyl monoglycidyl which can be used together, a compound which was illustrated below besides what is expressed with the above-mentioned general formula (10) can be used. Namely, N-substitution or N.N-substitution nonogarboxylic acid, such as cinnamic acid glycidyl and vinylbenzoic acid glycidyt, Styrene, Styrene 0051](Meta) As an acrylic-acid-akyl-ester monomer and an unsaturation group content monomer nonomers, such as alpha-methylstyrene and ohlorostyrene; Acrylonitrile, Oyano group content ester or diglycidyl ester of glycidyl ester, unsaturated dicarboxylic acid of unsaturation

propionate; diene system monomer, olefin; halogenation olefin; unsaturated estar; vinyl ether, such as Meta) It can polymerize by a radical polymerization, anionio polymerization, cationic polymerization, 0052]The manufacturing mathod in particular of the polymer (B) in this invention is not restricted. outadiene, isoprene, and chloroprene, etc. can be used.

nonomers, such as 2,4-dicyano butene-1; Vinyl acetate, Vinyl ester system monomers, such as vinyl

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alkyl-ester monomer as an essential ingredient. In this invention, as for a polymer (B), polymerizing in a radical polymerization may be preferred, and the gestalt may be any of solution polymerization, an etc., using the above-mentioned unsaturation group content monomer which uses an acrylic-acidamulation polymerization, suspension polymerization, and bulk polymerization.

0053]When carrying out a radical polymarization, a radical polymarization initiator is usually added as polymerization initiator, 2,2"–azobisisobutyronitrile, 2,2"–azobis 2-methylbutyronitrile, 2,2"–azobis (2,4nitiator is not necessarily required. It is preferred to perform the above-mentioned reaction at 20radical polymerization initiator which can be used in this invention, the polymerization initiator and metal compound catalyst of a peroxide system, azo, or a redox system are mentioned. As a radical dicarbonate, etc. can be illustrated. When activated with radiation or heat, a radical polymerization polymerizing under existence of an oxyalkylene polymer (A) is praferred. By polymerizing a polymer purposes, such as molecular weight control, and a chain transfer agent may be added. As a chain transfer agent, alkyl mercaptan, such as n-dodecyl mercaptan, t-dodecyl mercaptan, and n-butyl ngredient of this inventions other than a polymer (B), or may polymerize under existence. When a radical source of release to the above-mentioned unsaturation group content monomer. As a polymerizing undar existence of hardenability constituent ingredients other than a polymar (B). 0054]When compounding a polymer (B) by a radical polymerization in this invention, it is the (0055)A polymer (B) may polymerize under the nonexistence of the hardenability constituent dimethylvaleronitrile), benzoyl peroxide, t-butyl peroxide, acetyl peroxide, disopropyl peroxi 200 ** (preferably 50-150 **) for several hours - tens hours. mercaptan, a alpha-methylstyrene dimer, etc. can be used.

- SIX, R1 (3-a) - (1)

expressed with a following general formula (1).

formula (1), in particular the number and the existence part of a reactive silicon group in the molecule [0057]When a polymer (B) has a reactive silicon group expressed with the above-mentioned general the and or side chain of a polymer (B) molecule may be sufficient as an existence part, or they may of a polymer (B) ara not restricted. The number of a reactive silicon group is just one or more, and R1, X, and a are synonymous with said R1, X, and a among [type.] be both an end and a side chain.

general formula (1), following (i) - the method of (iv) are mentioned to a polymar (B). The method of (0058)As a method of introducing the reactiva silicon group axpressed with the above-mentioned the following (i) - (iv) may be combined and may be performed.

(iii) How to use the initiator which has a reactive silicon group expressed with a general formula (1) in (i) How to use together the unsaturation group content monomer which has a reactive silicon group (ii) How to use the chain transfer agent which has a reactive silicon group expressed with a general expressed with a general formula (1) in polymerizing an unsaturation group content monomer. formula (1) in polymerizing an unsaturation group content monomer.

polymerizing an unsaturation group content monomer.

0059]The compound which is used in the method of the above-mentioned (i) and which is expressed with a following general formula (11) as an unsaturation group content monomer which has a reactive unsaturation group content monomer uses together the unsaturation group content monomer which (iv) A method to which the compound which has a reactive silicon group which in polymerizing an has a specific functional group, and is expressed with the specific functional group concerned, a reactant basis, and a general formula (1) is made to react.

Re shows among [typa the univalent organic group which has an unsaturation group. R1, X, and a are R⁶-SiX₃R¹ (3-a) -(11)

silicon group expressed with a general formula (1) is preferred.

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'ynonymoue with said R1, X, and a.]

scrylcylcyspropyl trimethoxysilane and 3-methacrylcyl oxypropyl trimethoxysilane are preferred, the nertioned general formula (1) used in the method of the above-mentioned (ii), For example, gammaabove-mentioned compound comes out and requires using combining one sort or two sorts or more.)0061]As for the unsaturation group content monomer which has a reactive silicon group expressed nethyl dimethoxysilane, 3-aoryloyloxypropyl trimethoxysilana, (meth)aoryloyloxy silanes, such as 3exypropyl triethoxysilane, etc. can be mentioned. Especially in the above-mentioned compound, 3with the above-mentioned general formula (1), it is preferred to consider it as 0.01 - 20 mass part Vinyttrimetoxysilane, vinytriethoxysilane, vinyt trichloroeilene, Vinylsilane, such as tris (2-methoxyethoxy) vinylsilans: 3-soryloyloxypropylmethyldimethoxysilane, 3-methacryloyl oxypropyl icryloyloxypropyl triethoxysilane, 3-methacryloyl oxypropyl trimethoxysilane, and 3-methacryloyl propenyloxy silane, (CH₃O) Disulfide bond content compounds, such as 3Si-S-S-Si(OCH₃) 3 and nercaptopropyl triethoxysilane, gamma-mercapto propylmethyl diethoxysilane, The mercaptan 0062]As a chain transfer agent which has a reactive silicon group expressed with the abovecompound which has hydrolytic silyl groups, such as gamma-mercapto PUROPIRUTORIISO nercapto propyltrimethoxysilane, gamma-mercaptpropylmethyl dimethoxysilane, gamma-/inylmethyldimethoxyeilene, vinylmethyldiethoxysilane, vinyl methyldi chloroailicene, 0060]As a compound expressed with the above-mentioned general formula (11) among the total monomer 100 mass part used for composition of a polymer (B).

[0083]n [can use as an initiator the azo compound which has a neative elicon group expressed with the above mentioned gravitational (%) for the compound of being the above mentioned (%) for the enempte, and I the method of the above-mentioned (%). For example, and I the method of the above-mentioned (%). For example, the compound expressed with the polymer (B) which has an isocyanate group, and the above-mentioned general formula (5) can be CH₂₀₃Si-(CH₂) ₃-S-S-(CH₂) ₃-Si(OCH₃) ₃, are mentioned.

raise especially the extension characteristic of the hardenability constituent efter herdening. [0086]Next, the organic realn minute hollow body (O) in the hardenability constituent of this invention ody (C), an organic resin minute hollow body (C) may expand with heating etc. in this case and mean [0084]Adthough the molecular weight in particular of the polymer (B) explained above is not restricted, it is preferred as Mn that it is 500-100000, and it is more preferred that it is 2000-13000. of hardening, it becomes possible to raise intensity, weatherability, etc. of a hardenability constituent terminal by the method of being (ii) using the chain transfer agent which has a reactive silicon group. is explained. As for the organic rash minute hollow body (O) in this invention, tha mean particle diameter which the husks wall comprised with organic rashs says a spherical hollow body of 1 mm or micrometers is preferred, its 1-250 micrometers are more preferred, and its 5-100 micrometers are still more preferred. Although low molecular weight compounds, foaming agents, etc., such as low boiling point hydrocarbon, may exist in the inside of the husks wall of an organic resin minute hollow combination arises between the reactive silicon groups in a polyoxyalkylene polymer (A) at the time and the method of (iii) using the initiator which has a reactive silicon group, it becomes possible to When Mn of a polymer (B) exceeds 100000, workability tends to worsen, and when Mn is less than [0065]When a polymer (B) has a reactive silicon group expressed with a general formula (1), since after hardening. Since a polymer (B) with e reactive silicon group can be obtained to a molecular particle diameter may increase, the above-mentioned mean particle diameter means the mean ess. As for the mean particle diameter of an organic resin minute hollow body (C), 1-500 500, it is in the tendency which becomes insufficient [weatherability]. particle diameter before expansion.

[0087]in this invention, an usable organic resin minute hollow body (a minute hollow body is hereafter called "balloon") can be divided roughly into a thermosetting resin balloon and a thermoplastics oalloon. As a thermosetting resin balloon, a phenol resin balloon, an epoxy resin balloon. A urea resin palloon is mentioned and a polyvinylidene chloride system reain balloon, a polystyrene balloon, a colymethacrylate balloon, a polyvinyl alcohol balloon, and a styrene acrylic resin belloon are nentioned es e thermoplestics balloon.

[0083]in this invention, the balloon which covered the surface of the above-mentioned tharmopleatios balloon with thermosetting resin, and the thermopleatica belloon with chemostaturated the bridge can

balloon (called a seran balloon) in which a husks wall consists of polyvinylidene chloride system resin, and, as for polyvinylidene chloride system resin, refers to the homono/herrization hints of a variety described the copolymer of a varylidene chloride, the copolymer of a varylidene chloride, the copolymer of a varylidene chloride, are these mixtures intex. As monomore 3EADS provided by BASF A.G. As a constructed type styrene acrylic acid balloon of a bridge, SX863 P) provided by Japan Synthetic Rubber Co., Ltd. can be used, As a balloon which coated the surface 0069]As a phenol resin balloon, PHENOLIC MICROBALLOONS provided by UNION CARBIDE can be by EMERSON& CUMING by EMERSON & CUMING as a urea resin balloon as an apoxy ream balloon. 0070]As a polyvinylidene chloride eystem resin balloon, SARAN MICROSPHERES provided by DOW CHEMICAL, Expencel provided from Japanese phyllite, and the Matsumoto microsphere provided by used, for example. It is usable in ECCOSPHERES VF-O provided with ECCOSPHERES EP provided organic resin minute hollow body (C). The polyvirylidene chloride system resin balloon refers to the EXPANDABLE POLYSTYRENE provided by ARCO POLYMERS and EXPANDABLE POLYSTYRENE of the thermoplastics balloon with inorganic powder, such as calcium carbonate, talo, and titanium oxide, the Matsumoto microsphere MFL series provided by Matsumoto Yushi-Seiyaku can be used. 0071]In this invention, it is preferred to use a polyvinylidene chloride system resin balloon as an hermoplastics belloon with inorganic powder, such ae calcium carbonate, tale, and titanium oxide. also be used. It is usable also in the balloon which coated the surface of the above-mentioned other then the vinylidene obloride which constitutes the copolymer of a vinylidene obloride. Matsumoto Yushi-Seiyaku can be used. As a polystyrene balloon, it is usable in DYLITE

100 mass part, and it is preferred that it is especially 1 - 50 mass part. As for an organic resin minute [0072]The hardenability constituent of this invention is a thing containing the oxyalkylene polymer (A) and polymer (B) which were explained above, and an organic resin minute hollow body (G), Although such composition ratios in particular are not restricted, in this invention, it is preferred that a polymer oxyalkylene polymer (A) and a polymer (B), it is more preferred that it is 0.05 - 10 mass part, and it is (B) is 1 - 300 mass part to oxyalkylene polymer (A)100 mass part, it is more preferred that it is 1 nollow body (C), it is preferred that it is 0.01 - 20 mass part to a total of 100 mass parts of an (mete), the compound expressed with a general formula (10), etc. can be used, for exemple. preferred that it is especially 0.1 - 5 mass part.

VOM/PVC, acrylonitrile, the acrylic-acid-alkyl-ester monomer expressed with a general formula (9)

[0073]The hardenability constituent of this invention can be obtained by compounding a polymer (B) by an above-mentioned method apart from this oxyalkylene polymer (A), and mixing these and an organic resin minute hollow body (C), for example, after compounding an oxyalkylene polymer (A) by an above-mentioned method. After compounding an oxyelkylene polymer (A), it can obtain also by compounding a polymer (B) under existence of this polymer, adding an organic resin minute hollow existence of en oxyalkylene polymer (A) and an organic resin minute hollow body (C). An organic body (C) to thie, and mixing. In the case of the latter, a polymer (B) may be compounded under

oxyallylene polymer (A), e polymer (B), and en organio resin minuta hollow body (C), auch as a bulking caloium cerbonate with a mean particle diameter of 1-3 micrometers manufactured as a bulking agent mixing to remove this orgenic solvent es occasion demands. [0074]The herdenability constituent of this invention may contain additive components other than en 0075]First, the bulking agent which can be used for the hardenability constituent of this invention is segmentation method. The colloid calcium carbonate which carried out the surfece treatment of the explained. In this invention, a bulking agent can be added to a hardenability constituent. Precipitated solvent, a plesticizer, and an antiaging agent. The method in particular of adjusting the hardenability constituent containing such an additive component is not restricted, but should just add an additive component in once or several steps at the auitable stage after manufacture in the middle of agent, a hardening accelerator, an edhesive grant agent, e dehydrator, a thixotropy grant agent, a surface with fatty acid or a resin acid system organic matter, Calcium carbonate [, such as minor nature celcium cerbonata,]. fumed silics; -- sedimentation nature silics; -- surface siliconization solvent can be used in the cess of composition or mixing, and it is possible after composition or manufacture of a hardenability constituent. Hereafter, these additive components are explained. with heavy calcium carbonate with a mean particle diameter of 1-20 micrometers and a

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pentonite; — ferric oxide; -- zinc oxide; -- active white; -- resin beads and wood flour, pulp, e cotton

magnesium oarbonete; -- diatomite; -- celcination clay; -- clay, -- talc; -- titanium oxide; --

silica pulverized coal; -- silicio acid anhydride; -- hydrous-silicio-acids; -- carbon black; --

gent, glass fibers, such as farina, graphite, aluminum impalpable powder, and the Flint powder, a glass mics, and the blacking wash farina --- it rubs and fibrous fillers, such as powder state bulking lament, oarbon fiber, the Kevlar taxtiles, and a polyethylene fiber, are mentioned.

using a hollow body, the cobwebbing nature of a constituent can be improved and workability can be aised. Although a hollow body may be used independently, it may use combining the bulking agent of 0076]These bulking agents may be used independently and may use two or more sorts togother. In others, such as calcium carbonate. The amount of the bulking agent used in this invention has 1 -1000 preferred mass part to a total of 100 mass parts of an oxyalkylene polymer (A) and a polymer lardenability constituent and its hardened material can be carried out by using a hollow body. By hese, it is preferred to use calcium carbonate and especially the thing for which heavy calcium carbonate and colloid calcium carbonate are used together is preferred. The weight saving of a

nvention is explained. The crosslinking reaction of the reactive silicon group contained in the exyalkylene polymer (A) in this invention advances, even if the compound which promotes a reaction does not exist, but. When a reactive silicon group is alkoxy sllyl groups, for example, in order to make and a JIARUKIRUSUZU monoacetyl acetonate monoalkoxide, Tetravalent tin compounds, such as a dialkoxy tin mono- carboxylate, The Suzuki rate compounds, such as dialkyl tin bissoetylacetonate 0077]Next, the hardening accelerator which can be used for the hardenability constituent of this reactant of dialkyl tinoxide, the reactant of an ester compound and dialkyl tinoxide, and an alkoxy compounds, such as stearic acid tin; Dibutyltin dilaurate, Organic tin carboxylate like dialkyl tin dicarboxylates, such as dibutyltin discetate, dibutyltin mono- acetata, and dibutyltin malate, or [0078] As a hardening accelerator, 2-cthylhexanoio acid tin, naphthenic acid tin, Divalent tin practically sufficient cure rate reveal, it is preferred to use a hardening accelerator. B), and its 50 - 250 mass part is more preferred.

libutyitin monosoetyl scetonate monosikoxide, etc. are mentioned. The tin compound which carry out llisononyl ester, were made to react as a reactant of dialkyl tinoxide and an ester compound, and was made l'quefied is mentioned. In this case, as an ester compound, aliphatic series other than phthalic ester, ester of aromatic carboxylic acid, tetracthyl silicate, its partial hydrolysis condensate, etc. can se used. The compound which reacted or mixed these tin compounds with low molecule alkoxystlane 0079]As a Suzuki rate compound, dibutyitin bisacetylacetonate, dibutyitin bis-ethylscetoacetate, a heating mixing, and dibutyltin oxide and phthalic ester, such as dioctyl phthalate and phthalic acid silane compound and a dialkyl tin dialkyl sulfide, are mentioned.

sompounds, such as organic-carboxylic-acid bismuth salt; Phosphoric acid, p-toluenesulfonic acid, [0080]As a curing catalyst which can be used in addition to a tin compound, Divalent bismuth lexylamine, octylamine, decyl amine, lauryl amine, Aliphatio series monoamines, such as N.N-Acidic compounds, auch as phthalic acid and phosphoric acid di-2-ethylhexyl; A butylamine, etc. can also be used preferably.

enine compound, Amine compounds, such as aminosilane coupling agents, auch as alkanolamine, N-erminotetyN-C-eminoteropst intendencysilane, and 2-aminoteryOst, furthethosysilane, are mentioned. Since their landening feelilitatory effect, will improve it a division to momound and a division beautiful to As for the addition of the hardening accelerator in the case of using a hardening accelerator, it is preferred to consider it as 0.1 – 10 mass part to a total of 100 mass parts of an oxystkylene polymer above-mentioned hardening accelerator can also be used combining one sort or two sorts or more. riethylenetetramine, Aliphatic polyamine compounds, such as tetraethylenepentamine, an aromatic compound are used together with the 1st class amine compound, using together is preferred. The dimethyl- octylamine, athylanediamine, Hexamethylenediamine, disthylenetriamine,

agents, such as (meth)acryloyloxy group content Silang, amino group content Silang, sulfhydryl group constituent in order to improve an adhesive property. As an adhesive grant agent, silane coupling invention is explained. In this invention, an adhesive grant agent may be added to a hardenability 0082]As (meth)acryloyloxy group contant Silang, 3-methacryloyl oxypropyl trimethoxysilane, 3aoryloyloxypropyl trimathoxyailane, 3-methaoryloyl oxypropyl methyl dimethoxysilane, etc. are content Silang epoxy group content Silang, and carboxyl group content Silang, are mentioned. mentioned. As amino group content Silang, 3-aminopropyl trimethoxysilana, 3-aminopropyl triethoxysilana, 3-aminopropyl briethoxysilana, N-(2-aminopropyl)-3-aminopropyl

[0081]Next, the adhesive grant agent which can be used for the hardenability constituent of this

(A) and a polymer (B).

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głyciałyozy propylmettyl dinethozysilane, 3-głyciałyozy propyl triethozysilana, etc. are mentloned. As entrody group companie się przepiedne propylenie przepiedne się prze riethoxysilane, 3-mercapto propylmethyl dimethoxysilane, 3-mercapto propylmethyl diethoxysilane, rimethoxysilane, N-(2-aminoethyl)-3-aminopropyl methyl dimathoxyallana, N-(2-aminoethyl)-3minopropyl triethoxysilane, 3-ureido propyl triethoxysilane, N-(N-vinylbanzyi 2-aminoethyl)-3-0083]As aulfhydryl group contant Silang, 3-mercapto propyltrimethoxysllane, 3-mercaptopropyl stc. are mentioned. As spoxy group content Silang, 3-glycidyloxypropyl trimethoxysilane, 3uninopropyl trimethoxysilane, 3-anilino propyltrimethoxysilane, atc. are mentioned.

for 1 to 8 hours. The above-mentioned compound may be used slone and may use two or more kinds Silang, the reactant of epoxy group content Silang and suifhydryl group content Silang, the reactant nixing a silane coupling agent and agitating in a room temperature -150 ** temperature requirement together. The amount of the silane coupling agent used has 0 ~ 10 preferred mass part to a total of 0084] The reactant produced by making two or more sorts of silanc coupling agents react may be of suithydryl group content Silang, etc. are mentioned. These reactants can be easily obtained by used. As an example of a reactant, the reactant of amino group content Silang and epoxy group content Silang, the reactant of amino group content Silang and (meth)acryloxy group content

epoxy resin, various cyclosliphatic-epoxy-resin, N. and N-diglycidyl aniline, N.N-diglycidyl o-toluidine, triglycidyl isocyanurate, Epoxy resins, such as an epoxidation thing of unsaturation polymers, such as system epoxy resins, such as tetrahydrophtal acid digiycidyl and hexahydrophthalic acid digiycidyl, m [008]]n this invantion, an opcoy reain oan aleo be added as an adhesive grant agent. As an epoxy resimbly consultativent of this invention. A labelened A ediphoidy other types opcoy resim, this inhibit on the added to the hardenshildy delighted of their types opcoy resim, it is inhibited. Felighted of their type opcoy resim, it is interesting types opcoy. resins, such as a tatrabromobisphenol A-glyoldyl ether type epoxy resin, Novolak type epoxy resin, a propylene oxide addition, 4-glycidyloxy benzoic acid glycidyl, phthalic acid diglycidyl, Diglycidyl ester group, etc. are mentioned. The amount of [in the case used of adding an cpoxy resin] has 0 - 100 aminophenol series epoxy resin, a diaminodiphonylmethane system epoxy resin, Urethane modified hydrogenation bisphenol A type epoxy resin, The glycidyl ether type epoxy resin of bisphenol A / hydantoin type cpoxy resin, and petroleum resin. The vinyl system polymer containing an epoxy glycidyl ether of polyhydric alcohol, such as polyalkylene glycol diglycidyl ether and Blycerin, a 00 mass parts of an oxyalkylene polymar (A) and a polymer (B).

crossferred mass part to coyallydrae polymei (A)100 mass part. (1008)/flwen addite if the above poor resits to the Indentability constituent of this invention. the Indentability agent for curing catalyst) of an apoxy resit can be also be added further. As such a hardening pyromelitic anhydride,]; — phenoxy resin; — carboxylic acid; — alcoholic; — the oxyalkylene system spoxy group and it has at least one piece in intramolecular End carboxylation polyoxypropylene glycol suifone, Blocked amine; polyamide resir; imidateale compound disyandamide; boron trifluoride compounds, such as amine, auch as insophoronediamie and 2,45-tris/dimathyl aminomethyl phenol; bloce salts, or a kelonine compound; Phitalia suhydride, A hosahiydeophtalia danlydride, a plability — trings can be carried out. As such a dehydrator, ORUTOGI acid alkyla stratethyl acetato, such as methyl orthoformate and ethyl orthoformate. Altacetic acid alkyla, such as altethyl acetate; totrahydrophthalio anhydride, a dodecenyl succinic soid anhydride, carboxylic anhydride [, such as a ctc. are mentioned. The amount of [in the case used of using an epoxy resin hardener] has 0.1 to explained, in this invention, in order to improve the storage stability of a hardenability constituent, a etc.; The polybutediene, the hydrogenation polybutadiene, the acrylonitrile butadiene copolymer in Liquefied end functional group content polymers, such as an acrylic polymer, a ketimine compound [0087]Next, the dehydrator which can be used for the hardenability constituent of this invention is polymer (end amination polyoxypropylene glycol.) which averages the basis which can react to an little dehydrators are addad in the range which has an adverse effect on neither hardenability nor agent, triettyjenetetramine, tetraettyjanopantamine, Diettyjamino propylamine, N-aminoattyj piperazine, m-xylylene diamine, m-phenylenediamine, dlaminodiphenylmethane, dlaminodiphenyl which the end was embellished with a hydroxyl group, a carboxyl group, an amino group, etc., 300 preferred mass part to epoxy resin 100 mass part.

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hydrolytic organic silicon compound; hydrolytic organic titanium compounds, such as methyl

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container, such a dehydrator especially is affactive. As for the amount of tha dahydrator used in this tetraethoxyeilane are preferred. In the 1 liquid combination which adds a curing oatslyst etc. to tha mentioned. Especially, vinytrimetoxysilans from a point of a price and an effect and especially a nvention, it is proferred to consider it as 0.1 - 30 mass part to a total of 100 mass parts of an nardenability constituent of this invention, and is filled up with and used for a moisture-proof rimetoxysilane, vinyltrimetoxysilane, a tetramethoxy silane, and a tetraethoxysilane, etc. are

mentioned. As for a thixotropy grant agent, it is preferred to carry out 0.5-10 mass-part addition to a grant agent. As a thixotropy grant agent, hydrogenation castor oil, fatty acid amide, calcium stearate, constituent. The lappet nature of a hardenability constituent is improved by addition of a thixotropy 0088]Next, the thixotropy grant agent which can be used for the hardenability constituent of this nvention is explained. In this invention, a thix otropy grant agent can be added to a hardenability inc stearate, impalpable powder silica, organic acid processing calcium carbonate, etc. are oxyalkylene polymer (A) and a polymer (B).

explained, in this invention, a solvent can also be added to a hardenability constituent for the purpose aster, and ether are mentioned. By adding alcohol, the praenvation stability of the hardenability ocnstituent of this invention improves. Therefore, it is preferred to add alcohol, when carrying out the nothball of the hardenability constituent. As such alcohol, alkyl alcohol of the carbon numbers 1-10 of methanol, ethanol, isopropyl sloohol, isopentyl sloohol, hexyl sloohol, etc. is mentioned. As for a of adjustment of viscosity, and the improvement in preservation stability of a constituent. As this solvent, it is preferred to carry out 0.1-500 mass-part addition to a total of 100 mass parts of an solvent, aliphatic hydrocarbon, aromatic hydrocarbon, halogenated hydrocarbon, alcohol, ketone, 0089]Next, the solvent which can be used for the hardenability constituent of this invention is otal of 100 mass parts of an oxyalkylene polymer (A) and a polymer (B). oxyalkylene polymer (A) and a polymer (B).

polyester in which dibasic acid and dihydric alcohol are made to come to react. A polyether derivative 0091]Comparatively low-molecular plasticizers, auch as the above-mentionad phthalio ester, have a material surface or the adherend of the hardened material circumference may be produced, or it may olystyrene; oligomer, such as polybutadiena. Butadiene Aorylonitrila, polychloroprene, polyisoprene, arge plasticizing effect, in the hardened material of the hardenability constituent which uses these using such a low-molecular plasticizer, it is preferred to adjust content suitably in consideration of plasticizer, di-(2-ethylhexyl)phthalate, dibutyl phtalate, Phthalic ester, such as phthalic acid benzyl carboxylic-acid ester, pentaerythritol ester, such as a succinic acid screw (2-methylnonyl), dibutyl solyether, polyoxypropylene glycols, such as polyester plasticizer; polyoxyalkylene polyols, such as [0090]Next, the plasticizer which can be used for the hardenability constituent of this invention is plasticizer E. such as epoxy stearic acid benzyl.]; — chlorinated paraffin; — the hydroxyl group of constituent having an effect. Since the translatability to the surface of a plasticizer is high, when have an adverse effect also on the weatherability of the hardened material itself. Therefore, when explained. In this invention, a plasticizer can also be added to a hardenability constituent. As a butyl ester and phthalic acid diisononyl ester. Dioctyl adipate, Alcohol ester, such as aliphaticusing it as adhesives, an adhesive fall may pose a problem, and contamination of the hardened sebacate, and butyl oleate; Trioctyl phosphate, Phosphorio ester, such as tricresyl phosphate; Epoxidized soybean oil, 4,5-epoxy cyclohexane—1,2-dicarboxylic soid-di-2-ethyliexyl, epoxy which was closed by alkyl ethar, Polly alpha-methylstyrene, Oligomer of polystyrene, such as ow-molecular plasticizers while most generally being used from hypoviscosity-ization of a ookbutene, hydrogenation polybutane, and epoxidation polybutadiene, is mantionad. compatibility with a hardenability constituent, etc.

0092]in this invention, it is preferred that Mn uses what is called 1000 or more polymeric plasticizers aspecially as a hardening accalerator, after fixing by a compression state under a fixed condition, it is plasticizer may be used together, using only a polymeric plasticizer. By using a polymeric plasticizer, improvement in westharability. Epoxy plasticizera, auch as 4,5-epoxy cyclohoxane-1,2-dicarboxylic acid-di-2-ethylhexyl, When it is used combining divalent tin carboxylate and the 1st class amine reduction of the stain resistance on the surface of a paint, are acquired, and it contributes also to among the plasticizers illustrated above. In this case, a polymeric plasticizer and a low-molecular circumference stain resistance, drying improvement in the paint on a hardened material, and effects, such as reduction of the stain resistance of the surface of a hardened material or

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offective in a hardaned matarial with a returning large rate (comprassion recovery rate) when mmobilization is canceled being obtained. The above-mantioned plasticizer may be used

is usable in the compound of a hindered amine system, a benzotriazol system, a benzophenone series. antiaging agent, an antioxidant, an ultraviolet ray absorbent, light stabilizer, etc. are mentioned, and it nvention has 1 - 100 preferred mass part to a total of 100 mass parts of an oxyalkylene polymer (A) [0093]Next, the antiaging agent which can be used for the hardenability constituent of this invention is explained. In this invention, an antiaging agent can be added to a hardenability constituent. As an ndepandently or may use two or more sorts together. The amount of the plasticizer used in this and a polymer (B).

a benzoate system, a cyanoacrylate system, an acrylate system, a hindered phenol system, the Lynn

system, and sulfur systems, it is preferred to use it especially or more [of light stabilizer, an

entioxidant, and the ultraviolet ray absorbents I combining two. With such directions for use, antiaging ray absorbent, and the light stabilizer used, it is preferred respectively that it is 0.1 - 10 mass part to effects can be raised as a whole taking advantage of each feature. Especially the thing for which two a total of 100 mass parts of an oxyalkylene polymer (A) and a polymer (B), respectively. In less than antioxidant are specifically combined is effective. As for the amount of an antioxidant, an ultraviolet or more sorts chosen from the hindered amine light stabiliser of the 3rd class and the 2nd class, a benzotriazol system ultraviolet ray absorbent, a hindered phenol system, and a phosphite system 0.1 mass parts, when antiaging effects are not fully revealed and exceed tan masa parts, it is

ogether, although it may use independently is more preferred. In adding an air-oxidation hardenability compound, the amount used to a total of 100 mass parts of an oxyalkylene polymer (A) and a polymer to consider it as 0.1 - 50 mass part to a total of 100 mass parts of an oxyalkylene polymer (A) and a (B) 0.1 - 50 mass part, As for the amount used, when adding a photoresist compound, it is preferred constituent of this invention are explained. In this invention, it is possible to add an air-oxidation nardenability compound and a photoresist compound to a hardenability constituent. Adhesion of weatherability and dust is improved by adding these compounds. As for these compounds, using polymer (B). [0095]The alkyd resin produced by denaturalizing drying oil, such as tung oil and linseed oil, and 0094]Next, additive components other than the abova which can be used for the hardenability drying oil as an air-oxidation hardenability compound, The acrylic polymer, silicone resin, aconomically disadvantageous.

such as a polymer of diene of the carbon numbers 5-8 and a copolymer, and also these polymers, or polybutadione which denaturalized with drying oil, The denaturation thing of diene system polymers. 0096]Polyfunctional (mata) acrylate is mentioned as a photoresist compound, and as polyfunctional meta) acrylate Tetraethylena glycol diacrylate, pentaerythritol — doria — the acrylic ester (meta-) of polyhydrio aloohol, such as KURIRETO and trimethylolpropane triacrylate, can be illustrated. This photoresist compound is a monomer and a polymer (B) including an acrylio-acid-alkyl-eater (meta) a copolymer, a curing-in-air (mallein-ized denaturation, boiled oil denaturation, etc.) polyester compound, etc. are mentioned.

monomeric unit is distinguished.

simultaneously. Hexamethyldisilazane etc. can be used. The amount of [in the case used of using the constituent besides the above-mantioned compound. Aithough a hardenability constituent is colored hardenability constituent can also be added as a modulus regulator. By adding such a compound, the alcohol and phenol, etc. can be used and there is an effect which hardens late, so that the acidity of amine compound into a hardening accelerator especially is reduced, and stickiness of the surface is also reduced. As a compound which generates a trimethyl silanol, trimethylsilyl ether, such as fatty modulus of the hardened material at the time of making a divalent tin compound and the 1st class compound which generates a trimethyl silanol] has 0.1 ~ 10 preferred mass part to a total of 100 [0098]Organic colors, such as inorganic pigment; copper phthalocyanine blue, such as iron oxida, alcohol is strong. By changing the kind of alcohol arbitrarily, adjustment of hardenability is also possible and the trimethylsliyl ether of alcohol of the plurality for the purpose can also be used chrome oxide, and titanium oxide, and Phthalocyanine Green, can be added to a hardenability 0097]in this invention, the compound which generates a trimethy! silanol by hydrolysis in a by adding paints, the effect of improvement in weatherability is also expectable besides it. mass parts of an oxyalkylene polymer (A) and a polymer (B).

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0099]Surface appearance like a grante or grante can also be given by adding the microbody of the profession of the constituent, and a different clork to a tradenhability constituent in devict to give the stagm nature as sealant especially. It is also possible to add fire retardant, an artifungal agent, the

station merce as seattle subjector, at each possion or an or recommission memory assets the possion of the position of the po

The control sequence of the co

realized to go the used as a minute follow body. The theorems peniglic to rise the designer of a weight the Application of the properties of the properties of the properties of a minute hollow body. Then it is 1.p. 5-48525, which does not commiss or grain remain sense a minute hollow body. Then it is which contact increase, when the minute hollow body within consists of regarding reals it used to the grade of a weight averige bondoning familiaring the grade of a weight assign bonome bight also with a somewhat weight are also becoming familiaring the grade of a weight assign bonome bight also with a somewhat properties are also assigned that the properties of a possible as not necessarily also assigned the properties of the properties of the properties of the properties in not necessarily also assigned the properties of the properties of the properties of the properties of the analysis of the properties of a properties of the properties of the properties of a properties of a weight and the properties of a properti

outdoors to a rainstorm, sunlight, etc. for a long period of time, it can be used satisfactorily. Since the combination, and is combination of the moisture curing type which is kept where hygroscopic surface clear, either, an organic resin minute hollow body, Compared with minute hollow bodies, such as glass molature is intercepted, reacts to the molsture in tha air at the time of use, and is hardaned from the end it is assumed that this becomes insufficient [a weight saving]. [0102]The organic rasin minute hollow body in the hardenability constituent of this invention not only prevention and slump reduction in which achievement is difficult when minute hollow bodies, such as glass balloons, are used. Although the reason which this phenomenon produces is not necessarily [0104]When using for the above uses, the hardenability constituent of this invention can be made 1 surface. On the other hand, 2 liquid combination is the two-ingredient type of the base resin which uses a hardening accelerator as the main ingredients, and is combination of the reacting cure type balloons, to a polymer including an oxyalkylene polymer and/or (meta) an acrylic-acid-alkyl-ester 0103]Therefore, the hardenability constituent of this invention is suitably applicable as sealant, a degree of a weight saving is high, while being able to attain the weight saving of the subject which usea a hardened type oxyallylene polymer as the main ingredients, and the hardening agent which water blocking material, adhesives, a costing agent, etc., and even if it is a use which is exposed iquid combination or 2 liquid combination. 1 liquid combination is the one-ingredient type which enables improvement in a weight saving as mentionad above, but also enablas the cobwobbing monomeric unit, compatibility is high and having done the strong interaction more is assumed. contains a hardened type oxyalkylene polymer and a hardening accelerator during the same applied the hardenability constituent of this invention, it contributes also to a cost cut.

wherefull planes greated still find the standard security of the interrich herotricit, this invention herotricit is not find the these campbes, in the following complete of an interference, campbes, and required the campbes and the standard security of the standard secur

reacted and hardened by kneading these at the time of use.

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JP.2002-155201,A [DETAILED DESCRIPTION]

group at the end was obtained in the observation of branched manual LSS.

The configuration analysis and the observations of the configuration and the configuration of the communication of the configuration of the communication of the configuration of the communication of the configuration of the confi

company)3 copy, Added the tinuvin 327(made in Tiba Specialty Chemicals)1 copy as an ultraviolet ray 551DE (made by a Japanese phyllite company), Added 1.5 copies of N-(2-aminoethyl)-3-aminopropyl enough, and the hardenability constituent was obtained. * and a number were given to what used the sbbreviation etc. for the raw material of the hardensbility constituent in Table 1, and the details were 0109](Comparative example 1) Replaced with 0.6 copy of organic resin minute hollow body, and five nardenability constituent was produced like Example 1. The raw material and the presentation (mass [0106](Example 1) The hardenability constituent of Example 1 was produced by the raw material and company make.) 75 copies of formation CCR of white gloss, and heavy calcium carbonate (Shiroishi presentation (mass ratio) which are shown in following Table 1. namely, --- as opposed to Pa of 100 ethylhoxyl phthalate, As a thixotropy grant agent, DISUPARON #6500(made by Kusumoto industrial copies — as a bulking agent — fatty acid processing colloid calcium carbonate (Shiraishi industrial emperature with a 3 more paint roll, As an organic resin minute hollow body, 0.6 copy of Expanoel trimethoxysilane as an adhesive grant agent, three copies of vinyltrimetoxysilane was added as a Jehydrator, one copy of dibutyltin bisacetylacetonate was added as a curing catalyst, it kneaded copies of glass balloons (Tokai industrial company make, cell star Z-36) were used, and also the entioxidant, and it mixed uniformly. After carrying out kneeding mixing of this enough at a room calcium company make.) As 75 copies of HOWAITON SB, and a plasticizer, 40 copies of di-2specifient, the ADEKA stub LA62(made by Asahi Donka Kogyo K.K.)1 copy was edded as light stabilizer, the IRUGA NOx 1010(made in Tiba Specialty Chemicals)1 copy was added as an decompression and the polymer (henceforth "Pa") was obtained. ratio) were shown in Table 1. shown in Table 3.

(Comparative exemple: 2) An organic resul minute hollow body was not used, and size the Comparative constituent was produced like Example 1. The raw material and the presentation (mass result) were shown in Table 1.

[0110] [Table 1]

	28 競合体	大学が		東西に	子本ン性付本業	我少额政会出	亚州州州	操作的止射	物源性特多數	本人当	語の食具	報用を開	ACM. 7 . C. II.
					本本製	(1)		豪	4			有指的指数小中型体	1
	ě	自動化CCR**	ポワイトンSB7	pop**	ディスノキロン かちちのび	FXピン327%	アデカスタブしゅらど	412.25.2.10107	シラン化を整き	VTMS*	DBTDAA***	エクスパンセル551DE"	*N.7.9-2-36'U
	100	Ę	ĸ	9	3	-	-	,	1.5	3	-	9'0	-
-	100	75	ĸ	40	3	-	-	-	1,6		-	-	4
	100	75	Ę	9	8	-	-	-	1.5	9	-	1	

[0111]Applied the constituent of Exemple 1 and the comparative exemples 1-2 on the eluminum plete of 1-mm thickness so that it might be set to 5 mm in thickness, and recuperated oneself for seven of 1-mm thickness.

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wayou under 600 of humanizing at 20 % who for them chieful methods method in the most edit to form on said alianinan piles, and this was made after the specimen. The exposure assumination was done for this actions with the State and this was made after the specimens. One List and action is made and the state of the surface was the surface was of the surface was of the surface was of the surface was of the surface and the surface was of the surface of the surface was of the surface was of the surface of the surface of the surface was the surface of the surface of the surface was the surface of the s

And and coache which wave carefued cut planely we acceptate in the comparative assembles 1–2 on the aluminum (IDII (2)Next, agained the constituent of Exemple 1 and the comparative assembles 1 and the comparative assembles of 1 mm thickness as on that I might be set to 12 mm thickness, and includes 1 means to 5 mm of 10 mm thickness, and the comparative observed to 5 mm of 10 mm thickness, and the comparative of 10 mm thickness are set to 10 mm thickness. The set course assemble to 6 mm of 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thickness are set to 10 mm thickness are set to 10 mm thickness and 10 mm thickness are set to 10 mm thic

This quality design contains at the food of coundright constitution of the common is and the constitution of Extramol is and the conversal part of the contract of

and similar 1-2. This expeciping partly of the contributed feature including of Electropic I study the concentrative communities communities to comparative communities that the contribution of 18 KVIII.

This examely for including an examination is naturnized in the following table 2, and is anything when When this constitution that concernment is the includence in the includence in the contribution the constitution of the constitution of the constitution of the contribution of the

(I) 15th, the handonibility constituent of Esungle 1, a ligred was not characted to the ligent hand (I) 15th, the handonibility constituent of the constantial example and 2 is this almost neath him handonibility constituent of Esungle 1, although the amount of the facility and 2 is this almost neath handonibility constituent of Esungle 1, although the amount of the facility constituent of handonibility constituent of the second as the 2 is the second facility constituent of the second product of the second constituent of the constituent of the constituent of handonibility constituent of the comparative according to the constituent of handonibility constituent of the comparative according to the constituent of handonibility constituent of the comparative according to the constituent of handonibility constituent of the comparative according to the specific product of the constituent of handonibility constituent of

[0116] [Table 2]

			2		
			-	-	ď
の種類	整条体(Smm界)	10003489	0	0	o
100		150009 [8]	0	0	a
		190001	٥	0	a
	前機性(0.2mm厚)	30019101	0	0	٥
		800H	0	0	Ç
		S0096 [E]	0	0	O.
	第工徒(へら切れた		0	×	×
	スランプ(mm)		۰	~	"

[0117] [Table 3]

1.21 1.34 1.42

硬化物の比重

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1	14:3F
-	*1 国際基本国際経済要からから(自由日本社会)
*2	新賀次数カルシウム (自石カルシウム社数)
ee *	フタル酸ジー2ーエチルヘキシル
7	14 題称響フロア(著本合成社製)
	*5 ペンジドリアジール系術外線板技術 (25"Ax"3+974だMx"社類)
9 *	*6 ピンゲードアミン系光安定剤(結嘱化工業社職)
1.*	ヒンダードフェノール系数化防止剤 (タパスパクッラテイクミオルス゚社数)
* 8	*B N-(2-アミノエゲル)-3-アミノブロビルトリメトキシシラン
6*	ピールトリメトキシシラン
9.	*10 ジブチルスズピスアセチルアセトナート
=	*11 ポリ塩化ビニリテン系樹脂バルーン (日本スパ州社職、東比量:0.030)
* 12	- 12 - 11 2 11 11 1 1 1 1 1 1 1 1 1 1 1

[O118] The detection like a continuous detects according to this invention, a colorestating does not occur the thine of use, but the slump at the time of un-hardening is small, the weathershifty over the ting period of from etter intending is also consident, and it also enables the degree of a weight saving to provide a hardmanishy constituent high enough further.

Translation done.]